

Doped coupled frustrated spin- $\frac{1}{2}$ chains with four-spin exchange

Nicolas Laflorencie and Didier Poilblanc
*Laboratoire de Physique Théorique CNRS-FRE2603,
 , Université Paul Sabatier, F-31062 Toulouse, France*
 (Dated: February 1, 2008)

The role of various magnetic inter-chain couplings is investigated by numerical methods in doped frustrated quantum spin chains. A non-magnetic dopant introduced in a gapped spin chain releases a free spin- $\frac{1}{2}$ soliton. The formation of a local magnetic moment is analyzed in term of soliton confinement. A four-spin coupling which might originate from cyclic exchange is shown to produce such a confinement. Dopants on different chains experience an effective space-extended non-frustrating pairwise spin interaction.

PACS numbers: PACS numbers: 75.10.-b, 75.10.Jm, 75.40.Mg

Strong interest for quasi-one-dimensional frustrated spin chains has been revived since the discovery in 1993 of the first inorganic spin-Peierls (SP) compound CuGeO_3 which exhibits a transition [1] towards a gapped dimerised ground state (GS). Some vanadate oxides such as LiV_2O_5 are also excellent realizations of weakly interacting frustrated spin-1/2 chains [2]. Whether magneto-elastic couplings play a role at low temperature in LiV_2O_5 is not yet clear although it has been investigated theoretically [3]. Replacing a spin- $\frac{1}{2}$ in a *spontaneously* dimerised (isolated) spin chain by a non magnetic dopant (described as an inert site) liberates a free spin $\frac{1}{2}$, named a soliton, which do not bind to the dopant [4]. The soliton can be depicted as a single unpaired spin (domain) separating two dimer configurations [4]. The physical picture is completely different when a *static* bond dimerisation exists and produces an attractive potential between the soliton and the dopant [4, 5] and consequently leads, under doping, to the formation of local magnetic moments [4, 7] as well as a rapid suppression of the spin gap [6]. However, a coupling to a purely one-dimensional (1D) adiabatic lattice [8] does not produce confinement in contrast to more realistic models including an elastic inter-chain coupling (to mimic 2D or 3D lattices) [8, 9]. Doping quasi-1D dimerised spin chains is realized experimentally by substituting a small fraction of Copper atoms by Zinc (or Magnesium) atoms in CuGeO_3 [10].

The motivation of this Letter is to investigate by numerical methods based on Exact Diagonalisations (ED) of finite chains (see Ref. [9]) the role of various inter-chain magnetic couplings on soliton confinement. Such couplings are expected to become crucial in the case where magneto-elastic effects remain small. Indeed, in that case, we argue that the formation of magnetic moments in doped gapped frustrated spin chains is then primarily controlled by 4-spin interactions.

Let us first consider a model of coupled frustrated spin-

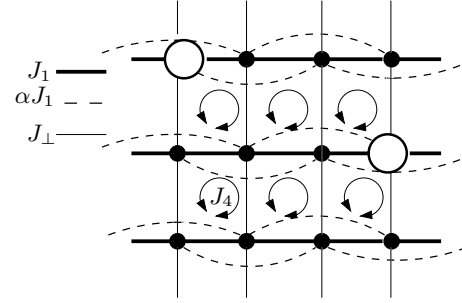


FIG. 1: Schematic picture of the coupled chains model with nearest neighbor, next-nearest neighbor, inter-chain and 4-spins couplings J_1 , $J_2 = \alpha J_1$, J_\perp , and J_4 . Full (resp. open) circles stand for spin- $\frac{1}{2}$ sites (resp. non-magnetic dopants).

$\frac{1}{2}$ antiferromagnetic (AF) chains, (see Fig.1)

$$H = \sum_{i,a} [J_1(\vec{S}_{i,a} \cdot \vec{S}_{i+1,a} + \alpha \vec{S}_{i,a} \cdot \vec{S}_{i+2,a}) + J_\perp \vec{S}_{i,a} \cdot \vec{S}_{i,a+1}], \quad (1)$$

where i is a lattice index along the chain of size L and a labels the M chains (L and M chosen to be even). Periodic boundary conditions will be assumed in *both directions*. In the absence of inter-chain coupling ($J_\perp = 0$) the behavior of the un-doped chain is controlled by the frustration α . From $\alpha = 0$ to $\alpha_c \simeq 0.241167$ [11], the chain is in a critical regime with a gapless excitation spectrum. Larger frustration stabilizes a spontaneously dimerised phase characterized by a gap opening. At $\alpha = 0.5$, the so-called Majumdar-Ghosh [12] (MG) point, the 2-fold degenerate Ground State (GS) consists in the product of singlets located either on odd or on even bonds. Beyond the MG point, the short-range correlations become incommensurate.

The inter-chain AF coupling J_\perp can stabilize a Néel ordered phase under some conditions. Following Schulz [13], a mean-field (MF) treatment of the inter-

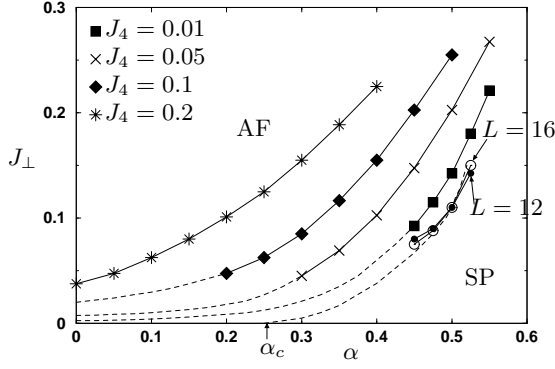


FIG. 2: SP-AF phase diagram in the (α, J_\perp) plane from ED of chains of length up to 16 sites. Symbols correspond to different values of $J_4 \geq 0$ as indicated on plot. Typically, FSE are smaller than the size of the symbols. The computed transition lines are extended by *tentative* transition lines (dashed lines) in the region where FSE become large. At $J_4 = 0$ we have plotted a few points in the vicinity of the MG point for $L = 12$ and $L = 16$.

chain coupling is performed,

$$H_{\text{eff}}(\alpha, J_\perp) = \sum_{i,a} [\vec{S}_{i,a} \cdot \vec{S}_{i+1,a} + \alpha \vec{S}_{i,a} \cdot \vec{S}_{i+2,a} + h_{i,a} S_{i,a}^z - J_\perp \langle S_{i,a}^z \rangle \langle S_{i,a+1}^z \rangle], \quad (2)$$

with

$$h_{i,a} = J_\perp (\langle S_{i,a+1}^z \rangle + \langle S_{i,a-1}^z \rangle), \quad (3)$$

the local magnetic field computed self-consistently. In the absence of dopant, we expect an homogeneous AF phase characterized by a self-consistent staggered magnetization $\langle S_{i,a}^z \rangle = (-1)^{i+a} m$ so that the system reduces to a single chain under a staggered magnetic field $h_i = \pm 2(-1)^i J_\perp m$. In the absence of frustration ($\alpha = 0$), it was shown that $m \sim \sqrt{J_\perp}$ [13]. By solving the self-consistency condition using ED of finite chains with up to 16 sites (supplemented by a finite size scaling analysis) the transition line $J_\perp = J_\perp^c(\alpha)$ (see Fig. 2) separating the dimerised SP phase ($m = 0$) and the AF ordered phase (for which $m \neq 0$) has been obtained in agreement with field theoretic approaches [14]. Finite size effects are small in the gapped regime and especially at the MG point ($J_\perp^c(\alpha = 0.5) \simeq 0.11$ for all sizes). Note also that numerical data suggest that the AF order sets up at arbitrary small coupling when $\alpha < \alpha_c$ with a clear finite size scaling $J_\perp^c(L) \propto 1/L$ at small α .

A non-magnetic dopant is described here as an inert site decoupled from its neighbors. Under doping the system becomes non-homogeneous so that we define a local mean staggered magnetization,

$$\mathcal{M}_{i,a}^{\text{stag}} = \frac{1}{4} (-1)^{i+a} (2 \langle S_{i,a}^z \rangle - \langle S_{i+1,a}^z \rangle - \langle S_{i-1,a}^z \rangle). \quad (4)$$

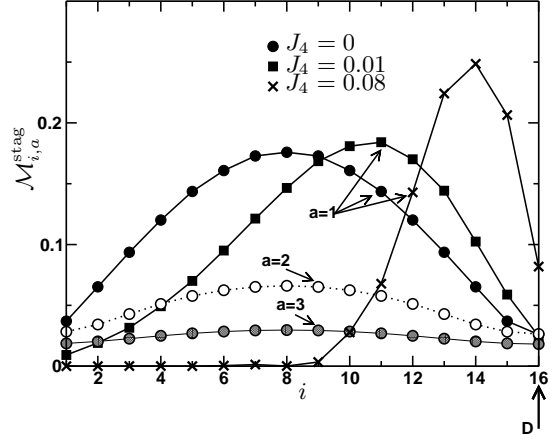


FIG. 3: Local magnetization $\mathcal{M}_{i,a}^{\text{stag}}$ for $L \times M = 16 \times 8$ coupled chains with one dopant D (shown by arrow) located at $a = 1$, $i = 16$ in the dimerised phase ($\alpha = 0.5$, $J_\perp = 0.1$). Circles correspond to $J_4 = 0$ (shown up to the third neighboring chain of the doped one) and squares (crosses) to $J_4 = 0.01$ ($J_4 = 0.08$). The coupling across the dopant has been set to 0 for convenience.

Following the method used in Ref. [9], the MF equations are solved self-consistently on finite $L \times M$ clusters and lead to a non-uniform solution. At each step of the MF iteration procedure, we use Lanczos ED techniques to treat *exactly* (although independently) the M *non-equivalent* finite chains and compute $\langle S_{i,a}^z \rangle$ for the next iteration step until convergence is eventually achieved.

We first consider the case of a single dopant. Whereas in the AF phase the magnetization profile is weakly enhanced in the vicinity of the dopant [15, 16, 17], in the SP gapped phase the soliton remains de-confined as can be seen from Fig.3. Note that the inter-chain coupling induces a "polarization cloud" with strong antiferromagnetic correlations in the neighbor chains of the doped one.

The fact that our treatment of J_\perp does not produce confinement is clear at the MG point where the dopant site separates two regions with out-of-phase dimer coverage. In such a state, the local magnetization $\langle S_i^z \rangle$ identically vanishes. Hence, we expect $J_\perp S_{i,a}^z \langle S_{i,a\pm 1}^z \rangle$ to be irrelevant. On the contrary, terms like $\langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle$ are 0 or $-3/4$ in the MG state and, hence, are relevant. Therefore, a 4-spin coupling $J_4(\vec{S}_{i,a} \cdot \vec{S}_{i+1,a})(\vec{S}_{i+1,a+1} \cdot \vec{S}_{i,a+1})$ which originates from cyclic exchange [18] is of crucial importance [19]. As done for J_\perp we have applied a mean-field treatment to the J_4 coupling leading to a (self-consistent) modulation of the intra-chain nearest neighbor couplings of the Halmiltonian,

$$H_{\text{eff}}(\alpha, J_\perp, J_4) = \sum_{i,a} [(1 + \delta J_{i,a}) \vec{S}_{i,a} \cdot \vec{S}_{i+1,a} + \alpha \vec{S}_{i,a} \cdot \vec{S}_{i+2,a} + h_{i,a} S_{i,a}^z + \text{constant}], \quad (5)$$

with $h_{i,a}$ given by Eq.(3) and

$$\delta J_{i,a} = J_4 \{ \langle \vec{S}_{i,a+1} \cdot \vec{S}_{i+1,a+1} \rangle + \langle \vec{S}_{i,a-1} \cdot \vec{S}_{i+1,a-1} \rangle \}. \quad (6)$$

As above, the numerical treatment again consists in a recursive diagonalisation of the M spin chains in the presence of the (non-uniform) MF magnetic fields *and* exchange modulations produced by its neighbors. Note that, as before, the effective coupling between the chains appears only through the MF self-consistency conditions so that, in practice, each chain Hamiltonian can be Lanczos diagonalised independently *at each iteration*.

Let us first consider the relative stability of the AF phase with respect to the dimerised phase in the undoped system with 4-spin coupling. Since all chains become equivalent we have to deal with an effective single dimerised frustrated chain problem in a staggered magnetic field. If the chains are not dimerised, $\delta J_{i,a}$ is constant and the exchange is just renormalised. On the other hand, if the SP phase is stable, each chain displays the same dimerised pattern when $J_4 < 0$ whereas dimer order is staggered in the transverse direction when $J_4 > 0$. Note that, apart from special features (see below), physical properties for positive or negative J_4 are quite similar. Note also that the dimerised GS would be 2^M -fold degenerate when $J_4 = 0$ (each chain is independently two-fold degenerate) while the degeneracy is reduced to 2 when $J_4 \neq 0$. When $\alpha = J_\perp = 0$ but $J_4 \neq 0$ each chain spontaneously dimerises and a gap opens up. Consequently, a finite value of the AF inter-chain coupling is necessary to drive the system into the AF ordered phase [20]. The frustration α stabilizes further the dimerised phase with respect to the AF one, the critical $J_\perp(\alpha)$ increasing with increasing α as seen from the phase diagram shown in Fig. 2. Note that finite size effects (FSE) become large at small J_4 and α and accurate extrapolations are not possible there.

Contrary to the previous case the spin- $\frac{1}{2}$ soliton released in a dimerised state by doping experiences a confining string which binds it to the dopant. Indeed, the J_4 interaction stabilizes (if one assumes an infinite number of chains) a given dimerisation pattern (over the two possible). Fig. 3 shows the enhancement of the staggered magnetization (4) in the doped chain in the vicinity of the dopant when $J_4 \neq 0$. Note that the dopant side where the soliton is bound is imposed by the (arbitrary) sign of the bulk dimerisation. A local spin-1/2 magnetization is then expected in the vicinity of the dopant.

In order to "measure" the strength of confinement, we define an averaged soliton-dopant distance as $\xi = \sum_i i |\langle S_i^z \rangle| / \sum_i |\langle S_i^z \rangle|$ so that $\xi = L/2$ in the absence of confinement ($J_4 = 0$) and ξ converges to a finite value otherwise when $L \rightarrow \infty$. On Fig.4, ξ is plotted versus J_4 for 2 different system lengths and $\alpha = 0.5$ and $J_\perp = 0.1$. FSE decrease for increasing J_4 . Note that $\xi(J_4) \neq \xi(-J_4)$ and a power law [5] with different exponents η is expected when $J_4 \rightarrow 0$. A fit gives $\eta \sim 0.33$

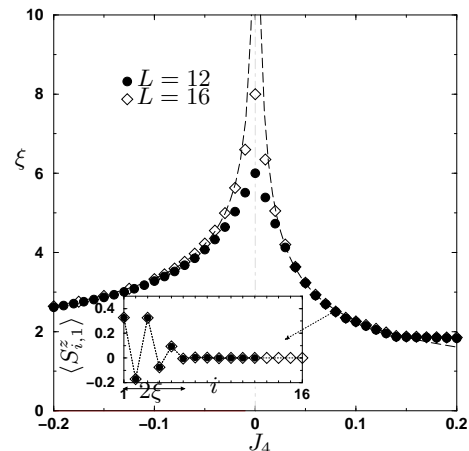


FIG. 4: ED data of the soliton average position vs J_4 calculated for $\alpha = 0.5$ and $J_\perp = 0.1$. Different symbols are used for $L \times M = 12 \times 6$ and 16×8 clusters. The long-dashed line is a power-law fit (see text). Inset shows the magnetization profile in the doped ($a = 1$) chain at $J_4 = 0.08$, ie $\xi \simeq 2.5$.

if $J_4 < 0$ and $\eta \sim 0.50$ for $J_4 > 0$ (Fig.4). This asymmetry can be understood from opposite renormalisations of J_1 for different signs of J_4 . Indeed, if $J_4 < 0$ then $\delta J_{i,a} > 0$ and the nearest neighbor MF exchange becomes larger than the bare one. Opposite effects are induced by $J_4 > 0$.

We now turn to the investigation of the effective interaction between dopants. We consider here a system of coupled chains with two dopants (see Fig. 1). Each dopant releases an effective spin $\frac{1}{2}$ which is localized at a distance $\sim \xi$ from it due to the confining potential set by J_4 . We define an effective pairwise interaction J^{eff} as the energy difference of the $S = 1$ and the $S = 0$ GS. When $J^{\text{eff}} = E(S = 1) - E(S = 0)$ is positive (negative) the spin interaction is AF (ferromagnetic). Let us first consider the case of two dopants in the same chain. (i) When the two vacancies are on the same sub-lattice the moments experience a very small ferromagnetic $J^{\text{eff}} < 0$ as seen in Fig. 5(A) so that the two effective spins $\frac{1}{2}$ are almost free. (ii) When the two vacancies sit on different sub-lattices, Δi is odd and the effective coupling is AF with a magnitude close to the singlet-triplet gap. Fig. 5(A) shows that the decay of J^{eff} with distance is in fact very slow for such a configuration. Physically, this result shows that a soliton and an anti-soliton on the same chain and different sublattices tend to recombine.

The behavior of the pairwise interaction of two dopants located on *different* chains ($\Delta a = 1, 2$) is shown on Fig. 5(B) & (C) for $J_4 > 0$. When dopants are on opposite sub-lattices the effective interaction is antiferromagnetic. At small dopant separation $J^{\text{eff}}(\Delta i)$ increases with the dopant separation as the overlap between the two AF clouds increases until $\Delta i \sim 2\xi$. For larger separation,

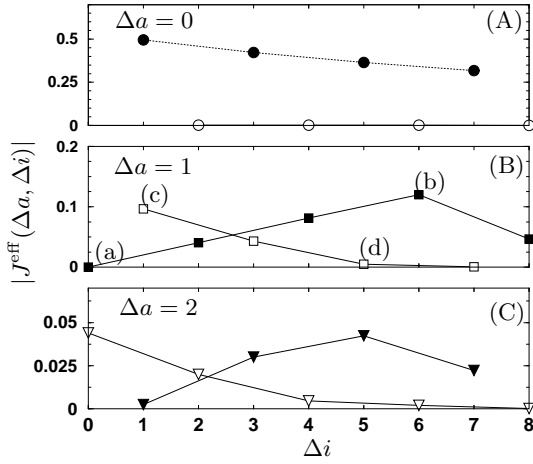


FIG. 5: Magnitude of the effective magnetic coupling between two impurities located either on the same chain (A) ($\Delta a = 0$) or on different ones (B) & (C) ($\Delta a = 1, 2$) vs the dopant separation Δi in a system of size $L \times M = 16 \times 8$ with $\alpha = 0.5$, $J_{\perp} = 0.1$, and $J_4 = 0.08$. Closed (resp. open) symbols correspond to AF (F) interactions.

$J^{\text{eff}}(\Delta i)$ decays rapidly. Note that the released spin- $\frac{1}{2}$ solitons bind on the opposite right and left sides of the dopants as imposed by the the bulk dimerisation [21]. If dopants are on the same sub-lattice, solitons are located on the same side of the dopants [22] and the effective exchange $J^{\text{eff}}(\Delta i)$ is ferromagnetic and decays rapidly to become negligible when $\Delta i > 2\xi$. The behavior of the GS local staggered magnetization is plotted on Fig. 6 for parameters corresponding to 4 typical behaviors. The key result here is the fact that the effective pairwise interaction is *not* frustrating (because of its sign alternation with distance) although frustration is present in the microscopic underlying model. AF ordering is then expected (at $T = 0$) as seen for a related system of coupled Spin-Peierls chains [9].

We finish with a discussion on the relevance to materials. Coexistence of AF ordering and SP order observed in Zinc-doped CuGeO_3 [10] can be understood as an ordering of the induced $S = 1/2$ moments in the dimerised background [9]. We have shown that a 4-spin magnetic inter-chain interaction, similarly to the coupling to a 2D (or 3D) adiabatic lattice [9] but in contrast to a pairwise inter-chain coupling, leads to the formation of local magnetic moments under doping by inert atoms. Intermediate-range *non-frustrated* spin exchange interactions between these moments are expected to stabilize a low-temperature AF phase. Therefore, cyclic exchange is expected to play a key role in quasi-1D frustrated spin chains like LiV_2O_5 [2].

We gratefully acknowledge stimulating discussions with I. Affleck and E. Sorensen.

Note added.—An additional small confinement $\propto J_{\perp}^2/J_1$

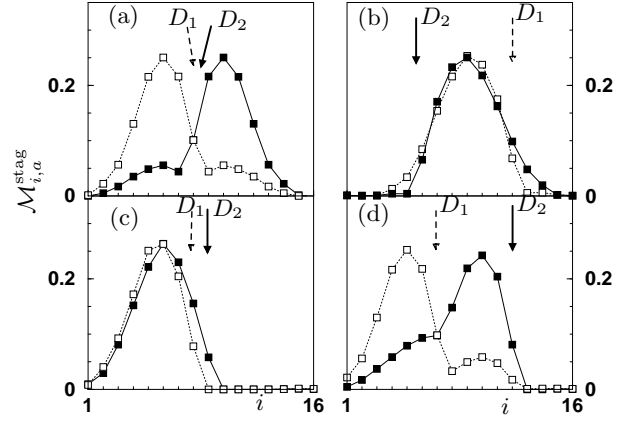


FIG. 6: Local staggered magnetization $\mathcal{M}_{i,a}^{\text{stag}}$ for 2 dopants (D_1 & D_2) on neighboring chains. The labels (a)-(d) refer to the data points shown on Fig. 5(B) corresponding to different relative positions of the dopants. Arrows stand for the dopant positions in the chain direction and full (open) symbols correspond to the chain doped with D_2 (D_1).

was found in second order perturbation in J_{\perp} [23].

-
- [1] M. Hase, I. Terasaki, and K. Uchinokura, Phys. Rev. Lett. **70**, 3651 (1993).
 - [2] M. Isobe and Y. Ueda, J. Phys. Soc. Jpn. **65**, 3142 (1996); R. Valenti et al., Phys. Rev. Lett. **86**, 5381 (2001).
 - [3] F. Becca, F. Mila and D. Poilblanc, cond-mat/0209278.
 - [4] E. S. Sørensen, I. Affleck, D. Augier, and D. Poilblanc, Phys. Rev. B. **58**, R14701 (1998).
 - [5] T. Nakamura, Phys. Rev. B **59**, R6589 (1999).
 - [6] G. B. Martins, E. Dagotto and J. Riera, Phys. Rev. B. **54**, 16032 (1996).
 - [7] B. Normand and F. Mila, Phys. Rev. B. **65**, 104411 (2002).
 - [8] P. Hansen, D. Augier, J. Riera, and D. Poilblanc, Phys. Rev. B. **59**, 13557 (1999).
 - [9] A. Dobry et al., Phys. Rev. B. **60**, 4065 (1999).
 - [10] M. Hase et al., Phys. Rev. Lett. **71**, 4059 (1993); S.B. Os-
eroff et al., Phys. Rev. Lett. **74**, 1450 (1995); L.-P. Reg-
nault et al., Europhys. Lett. **32** 579 (1995); T. Masuda
et al., Phys. Rev. Lett. **80**, 4566 (1998); B. Grenier et
al., Phys. Rev. B **58**, 8202 (1998).
 - [11] S. Eggert, Phys. Rev. B. **54**, R9612 (1996).
 - [12] C. K. Majumdar and D. K. Ghosh, J. Math. Phys. **10**,
1399 (1969).
 - [13] H. J. Schulz, Phys. Rev. Lett. **77**, 2790 (1996); see also
A.W. Sandvik, Phys. Rev. Lett. **83**, 3069 (1999).
 - [14] H. Fukuyama, T. Tanimoto and M. Saito,
J. Phys. Soc. Jpn. **65**, 1183 (1996).
 - [15] M. Laukamp et al., Phys. Rev. B. **57**, 10755 (1998).
 - [16] Similar effects are seen in the two-dimensional antiferro-
magnet; see e.g. N. Bulut, D. Hone, D. J. Scalapino, and
E. Y. Loh, Phys. Rev. Lett. **62**, 2192 (1989).
 - [17] For studies of quasi-1D antiferromagnets containing non-
magnetic dopants see e.g. S. Eggert, I. Affleck, and

- M. D. P. Horton, Phys. Rev. Lett. **89**, 047202 (2002).
- [18] Two-leg ladders with cyclic exchange show a very rich phase diagram. See e.g. S. Brehmer et al., Phys. Rev. B. **60**, 329 (1999); M. Muller, T. Vekua and H.-J. Mikeska, Phys. Rev. B **66**, 134423 (2002); A. Läuchli, G. Schmid and M. Troyer, cond-mat/0206153.
- [19] Our definition of J_1 includes the small contribution from cyclic exchange. We have shown that the other bilinear terms produced by cyclic exchange have negligible effects.
- [20] For numerical studies on anisotropic dimerized square lattice see M. Matsumoto, C. Yasuda, S. Todo, and H. Takayama, Phys. Rev. B. **65**, 014407 (2002) and references therein.
- [21] In that case, the solution is unique and the sign of the bulk dimerisation is fixed by the local condition of maximum overlap between the 2 solitonic clouds.
- [22] Two equivalent symmetric solutions are obtained with opposite signs of the bulk dimerisation.
- [23] T. M. R. Byrnes et al, Phys. Rev. B **60**, 4057 (1999).